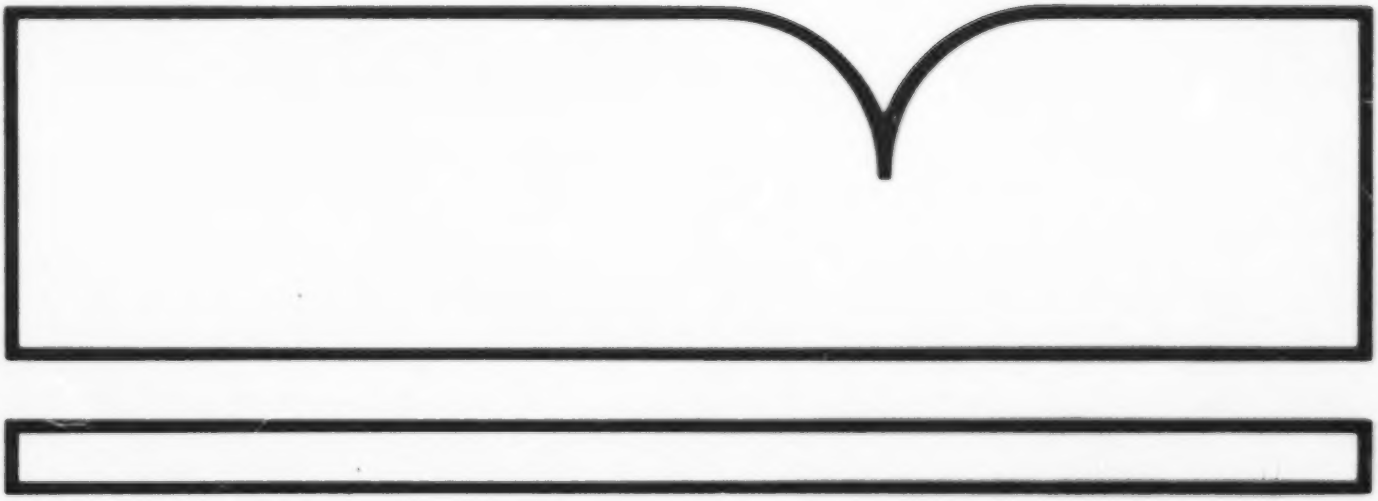
PB87-182648

FGD (Flue Gas Desulfurization), SCR (Selective-Catalytic-Reduction) Gain Coal-Fired-Boiler Experience in Japan

(U.S.) Environmental Protection Agency
Research Triangle Park, NC

1987



U.S. Department of Commerce
National Technical Information Service
NTIS

TECHNICAL REPORT DATA
(Please read Instructions on the reverse before completing)

1. REPORT NO. EPA/600/M-87/014	2.
3. REC	PB87-182648
4. TITLE AND SUBTITLE FGD, SCR Gain Coal-Fired-Boiler Experience in Japan	5. REPORT DATE 1987
	6. PERFORMING ORGANIZATION CODE
7. AUTHOR(S) Jumpei Ando (Chuo University) and Charles B. Sedman (EPA/AEERL)	8. PERFORMING ORGANIZATION REPORT NO.
9. PERFORMING ORGANIZATION NAME AND ADDRESS Chuo University Tokyo, Japan	10. PROGRAM ELEMENT NO.
	11. CONTRACT/GRANT NO. EPA Purchase Order 6D1999NAFX
12. SPONSORING AGENCY NAME AND ADDRESS EPA, Office of Research and Development Air and Energy Engineering Research Laboratory Research Triangle Park, NC 27711	13. TYPE OF REPORT AND PERIOD COVERED Journal Article; 12/86
	14. SPONSORING AGENCY CODE EPA/600/13

15. SUPPLEMENTARY NOTES AEERL project officer is Charles B. Sedman, Mail Drop 4, 919/541-7700. In Power, 131:2, pp. 33-36 (2/87).

16. ABSTRACT The paper discusses flue gas sulfur dioxide (SO2) and nitrogen oxide (NOx) control technology in Japan, where success in reducing SO2 and NOx emissions in recent years has been quite remarkable considering Japan's economic growth and the increased role of coal in Japanese energy production. Yearly SO2 emissions in Japan were reduced from 5 million tons in 1967 to 1.2 million tons in 1985, while NOx emissions were reduced from 2 million tons in 1973 to 1.3 million tons in 1985. Conversely, Japanese energy use increased from about 3.0 x 10 to the 15th power kcal in 1970 to 4.0 x 10 to the 15th power kcal in 1985, during which time coal has become increasingly important. Fuel coal imports, which began in 1977, reached 20 million tons in 1985, and are expected to increase to 50 million tons by 1990. Contributions to this success include: increases in hydro and nuclear energy production, lowering the sulfur in fuel oil via importing lower sulfur crudes and extensive oil desulfurization, and (most important) the development and successful implementation of SO2 and NOx controls. Since 1968, nearly 1500 flue gas desulfurization (FGD) systems have been constructed at roughly 45,000 MWe equivalent capacity. During the same period, more than 200 selective catalytic reduction (SCR) units for NOx control have been constructed at 35,000 MWe equivalent capacity.

17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group
Pollution Flue Gases Sulfur Oxides Nitrogen Oxides Coal Combustion	Pollution Control Stationary Sources Japan	13B 21B 07B 21D

REPRODUCED BY
U.S. DEPARTMENT OF COMMERCE
NATIONAL TECHNICAL
INFORMATION SERVICE
SPRINGFIELD, VA 22161

18. DISTRIBUTION STATEMENT Release to Public	19. SECURITY CLASS (This Report) Unclassified	21. NO. OF PAGES 5
	20. SECURITY CLASS (This page) Unclassified	22. PRICE

EPA Form 2220-1 (9-73)

EPA/600/J-87/014

FGD, SCR gain coal-fired-boiler experience in Japan

For years, Japan has reported its favorable experience with flue-gas-desulfurization and selective-catalytic-reduction systems—most of it on oil-fired boilers. Here's a glimpse of the experience with coal

By Jumpei Ando, Chuo University, and Charles B Sedman, EPA

Japan's success in reducing SO_2 and NO_x emissions in recent years is notable considering its economic growth and the increased use of coal in Japanese powerplants. Yearly SO_2 emissions have been cut from 5-million tons in 1967 to 1.2-million tons in 1985; NO_x emissions have been reduced from 2-million tons in 1973 to 1.3-million tons in 1985. Fuel-coal imports, beginning in 1977, reached 20-million tons in 1985 and are expected to soar to 50-million tons by 1990.

Development and implementation of SO_2- and NO_x-control systems have, in large measure, been responsible for the impressive reductions, although increased use of hydro and nuclear energy sources and low-sulfur fuel oil have helped as well. Approximately 45,000 MW of capacity, counting oil- and gas-fired plants, is being served by 1500 flue-gas desulfurization (FGD) systems installed since 1968. During the same period, more than 200 selective-catalytic-reduction (SCR) systems have been built to serve 35,000 MW equivalent capacity, including some combined-cycle installations (see box, next page).

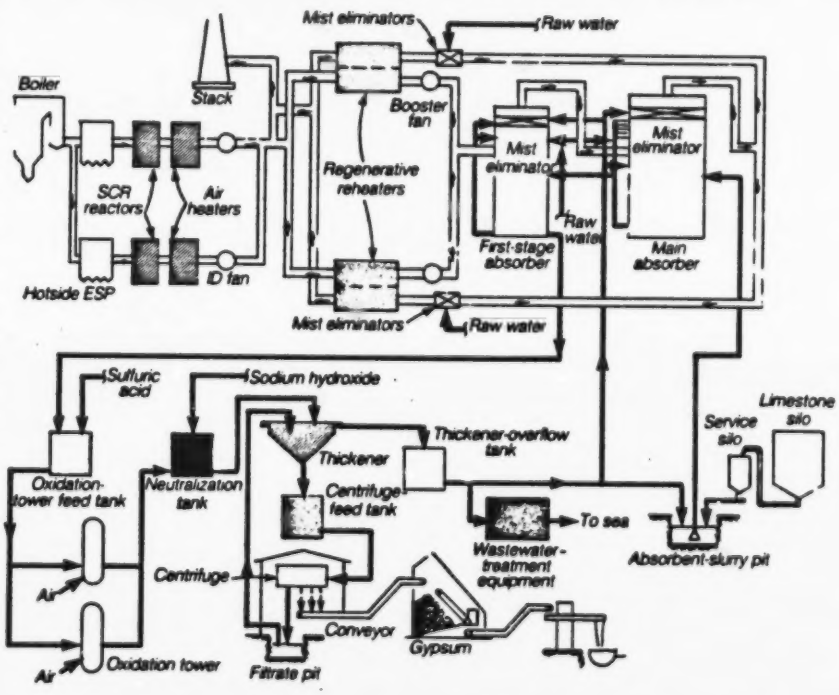
Developments in FGD and SCR technology in Japan bear watching. Combined with developments in Europe (POWER, February 1986, p 29), they are sure to be a factor in the US, especially if acid-rain legislation comes from Congress.

Coal-fired FGD overview

There are 49 coal-fired utility boilers in Japan, including those to be completed by the end of 1987. Eleven of the smallest boilers have neither FGD nor SCR and burn a domestic coal with 0.2% to 0.3% sulfur. Eleven larger boilers, comprising 3300 MW of capacity, have FGD units without SCR. And 27 boilers, representing 12,190 MW or 63% of the coal-fired capacity, have both FGD and SCR systems.

Nearly all coal-fired boilers in Japan employ wet limestone gypsum-producing FGD systems (POWER, February 1983, p 43). Except for a brief period in the mid-1970s, demand for gypsum in Japan has exceeded supply. Therefore, there are strong market incentives to produce gypsum of high quality.

Gypsum-producing processes for coal-fired utility boilers are characterized according to type of sorbent (calcium hydroxide or calcium carbonate) and additive (sodium- or magnesium-based) as well as the use of sulfuric acid for pH con-



1. Flue-gas-desulfurization system used in Japan produces salable gypsum, features sulfuric acid addition during oxidation step, hot-side ESPs and SCR reactor upstream.

NTIS is authorized to reproduce and sell this report. Permission for further reproduction must be obtained from the copyright owner.

trol and prescrubbers. Notably absent are sludge-producing systems common in the US. Because of a lack of space in Japan for sludge disposal, only one waste-sludge system has been installed.

The most popular system incorporates a prescrubber to remove chlorides and fluorides, collect remaining flyash, and cool the gas. Note that the prescrubber adds to system cost and itself produces a waste liquid and a sludge. Sulfuric acid is added to the main scrubber tower to help control pH. This system is also characterized by a separate oxidation step.

Of the eight most recent units (Table 1), five are of the type described above. The other three are characterized by sulfuric acid addition and use of a sodium promoter.

Other processes in use are variations of the above. For example, three plants in Japan have an additional scrubber vessel to lower the pH by further SO_2 absorption, and add sodium hydroxide or sulfate to promote absorption and oxidation. In another process, SO_2 absorption and oxidation of sulfite to sulfate take place in the same reactor.

Table 1: FGD and SCR units completed since 1977 for coal-fired utility boilers in Japan

Utility	Station name	Boiler capacity, MW	FGD-unit constructor[1]	FGD-unit startup date	SCR-unit constructor[1]	SCR-unit startup date	SCR-unit type[2]
Electric Power Development Co	Takehara	250	BHK	1977	BHK, KHI	1980	L
		700	IHI	1983	BHK	1983	L
	Ishikawa	125	IHI	1986	—	—	—
		125	IHI	1987	—	—	—
	Matsushima	2 × 500	BHK, IHI	1981	—	—	—
Chugoku Electric Power Co	Shimonoseki	175	MHI	1979	MHI	1980	H
		2 × 75	MHI	1982	MHI	1982	H
	Shin-Ube	156	MHI	1982	MHI	1982	H
		125,156	BHK	1984	BHK	1984	H
	Mizushima	500	IHI	1986	IHI	1986	H
	Shin-Onoda	500	IHI	1987	IHI	1987	H
Hokkaido Electric Power Co	Tomato	350[3]	BHK	1980	BHK	1980	L
		600	MHI	1985	—	—	—
Kyushu Electric Power Co	Omura	156	MHI	1982	—	—	—
	Minato	156	MME	1983	MHI	1983	H
	Matsuura	700	MHI	1987	MHI	1987	H
Joban Joint Power Co	Nakaso[4]	600	MHI	1983	MHI	1983	H
		600	MHI	1983	IHI	1983	H
Shikoku Electric Power Co	Saijo	156	KHI	1983	MHI	1983	H
		250	KHI	1984	IHI	1984	H
Tohoku Electric Power Co	Sendai	2 × 175	BHK	1983	BHK	1983	H
Toyama Joint Power Co	Toyama	2 × 200	CHI	1984	BHK	1984	H
Sakata Joint Power Co	Sakata	350	MHI	1984	MHI	1984	H
		350	MHI	1987	MHI	1987	H
Tokyo Electric Power Co	Yokosuka[5]	265	MHI	1984	MHI	1984	H
		265	BHK	1985	MHI	1985	H

[1]BHK—Babcock Hitachi K.K.; MHI—Mitsubishi Heavy Industries Ltd; IHI—Ishikawajima-Harima Heavy Industries Co Ltd; MME—Mitsui Miike Engineering Co Ltd; KHI—Kawasaki Heavy Industries Ltd; CHI—Chiyoda Chemical Engineering & Construction Co Ltd [2]L-Low-dust system H-High-dust system [3]Subjecting 50% of flue gas to FGD and 25% to SCR [4]Burns both coal and oil [5]Burns a coal/oil mixture

Table 2: Performance-test vs design data for the Takehara wet limestone FGD unit

	Design	Test operation
Boiler load, MW	700	700
Inlet SO_2 content, ppm	1000	505
Outlet SO_2 content, ppm	Less than 100	34
SO_2-removal efficiency, %	Greater than 90	93.3
Free moisture in gypsum product, % wt	Less than 10	8.65
Gypsum purity, % wt	Greater than 95	98.1
Calcium carbonate usage (98.9% purity), tons/hr	9.5	4.7
Sulfuric acid usage (98% purity), tons/hr	0.45	0.06
Caustic soda usage (25% concentration), tons/hr	0.49	0.04
Byproduct gypsum, tons/hr	18.5	8.2
Industrial-water usage, tons/hr	112.2	115.0
Power consumption, MW	17.4	14.0

All plants constructed since 1977 have electrostatic precipitators, removing 97-99.6% of the flyash. The scrubbers remove between 90% and 98% of the SO_2 as well as 70-90% of the remaining particulate matter.

Representative unit

The FGD plant for a 700-MW coal-fired boiler at Electric Power Development Corp's Takehara station uses the process depicted in Fig 1. It was chosen because very efficient hot-side ESPs could be installed to keep dust content low at the scrubber inlet. This results in less flyash contamination of the gypsum product without having a prescrubber loop. In addition, the small amount of sludge produced in a prescrubber could not be conveniently disposed of at this plant.

In the Takehara FGD plant (Fig 1), flue gas is treated by two trains of hot-side ESPs and SCR reactors and then passes through two regenerative reheaters arranged in parallel. A fan is located between the reheater and scrubber and a scavenging fan is added to prevent leakage of the untreated gas into the cleaned gas. Flue gas is introduced into the first scrubber with a two-stage spray, and from there goes into the main scrubber with a six-stage spray. Cleaned gas flows to a mist eliminator and finally through the reheater on out to the stack.

Limestone slurry is fed into the main scrubber and a portion of the circulating slurry in the main scrubber is fed into the first scrubber. Calcium sulfite slurry leaving the first scrubber is acidified with a small amount of sulfuric acid, then oxidized with air bubbles in the oxidizer. Gypsum is separated in a thickener, then further dewatered in a centrifuge.

Table 2 summarizes some of the pertinent performance data for the Takehara FGD plant and compares them to design. After 2100 hours of operation, the plant was shut down and inspected. Water containing soft deposits was found in the duct between the reheater and the first scrubber, but not enough to hinder plant operation. Some scale, less than a millimeter thick, was found at the top of the mist eliminator in the first scrubber. Some changes to the mist-eliminator washing scheme are expected to correct even minor scale buildup in this area.

Power consumption has been relatively low, averaging 2.0-2.1% of the power generated in the boiler.

A simplified version of the process used at Takehara has recently been installed at 156- and 47.5-MW plants. Essentially, it makes use of only one scrubber vessel instead of two (Fig 2). About 95% of the SO_2 is absorbed by the limestone slurry, which leaves the scrubber at a pH of 5.5. After a small amount of sulfuric acid is added to lower pH, the slurry is oxidized

Local ordinances tighten emissions standards further

It is well known that ambient NO_x- and SO_2-emissions standards set by the federal government are stringent in Japan. What is not as well known is that agreements between industry and local authorities and/or citizens groups have made these standards even more strict—and harder to meet with existing pollution-control technologies. To illustrate: While the emissions standard for NO_x is 400 ppm, local agreements have forced at least six utility boilers to meet an emissions limit that ranges from 160 to 300 ppm, depending on the locality.

On top of this, Japan has a program of giving aid to designated air-pollution patients, individuals who live in certain SO_2-pollution regions and are diagnosed as being victims of air pollution.

The number of these patients has increased ten fold since the early 1970s, despite improvements in air quality. Expansion of the regions in the program to include most sections of large cities—like Tokyo and Osaka—is responsible for much of this increase. Today, many groups are claiming that NO_x emissions are at least partially to blame, in addition to SO_2 emissions.

Since 1974, levies have been applied to SO_2 sources that emit more than 1.5 MW equivalent of exhaust gas. Owners of SO_2 sources must pay according to the amount of SO_2 emitted, even if the source is in compliance with regulations. The money is used to aid the designated air-pollution patients.

The SO_2-levy program has prompted the construction of hundreds of small FGD plants on boilers between 1 and 10 MW in size. According to some, they are not very cost-effective and often are operated without sufficient control. Other boiler owners construct new, more efficient FGD plants or increase the SO_2-removal capabilities of the existing FGD plant to reduce levies imposed by localities.

On one side of the controversy, some groups want to remove from the program those regions where air quality has improved substantially—and thus reduce the number of designated patients. On the other side are groups who claim that the patients' health problems are caused by NO_x and want to establish a levy program for NO_x emissions as well.

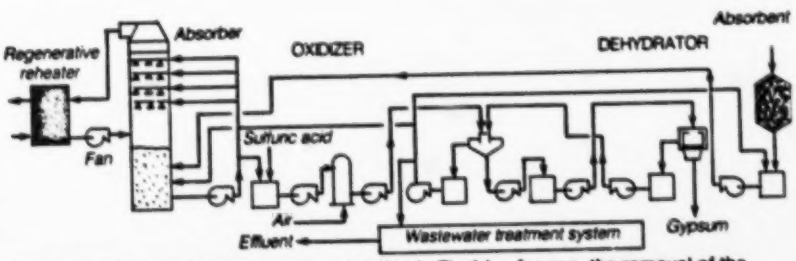
for complete conversion of sulfite to sulfate, or gypsum. The gypsum is then thickened and centrifuged to give a product containing less than 10% moisture.

The circulating liquor contains a small amount of sodium which serves as a catalyst or promoter of SO_2 absorption and sulfite oxidation. After wastewater treatment, a portion of the liquor is purged to prevent accumulation of impurities, such as chlorides. Sodium sulfate or sodium hydroxide is added to make up the lost sodium values.

Operating data from these two plants indicate that very high SO_2-removal and oxidation rates can be achieved with a single absorber when sodium is added. The high oxidation ratio may also result from the relatively low inlet SO_2 concentration. Elimination of the oxidizing tower and sulfuric acid addition makes the overall process more economical.

NO_x control

Most powerplants in Japan meet current NO_x regulations through combustion modifications. However, essentially all new coal-fired boilers must install post-combustion NO_x controls because of local agreements (see box) and federal standards that have recently been tightened. The most advanced post-combustion NO_x-reduction technique is selective catalytic reduction (POWER, August 1986, p 53 and October 1986, p 59). Table 3 shows some performance characteristics for several coal-fired utility boilers employing SCR. They typically have parallel-flow catalysts with space velocities in the 2000-4000/hr range to remove 60-80% of the NO_x. Space velocity for a coal-fired-boiler application is usually lower than for oil-fired units because the inlet



2. FGD system is simplified compared to that in Fig 1 by, for one, the removal of the first-stage absorber. Note treatment of effluent and gypsum-purification subsystem

Combined-cycle plants have SCR for NO_x reduction

At least eight combined-cycle power stations ranging from 144 to 2000 MW of capacity are, or will be, equipped with SCR as a result of local regulations. In most cases, these plants already reduce NO_x emissions in the gas-turbine exhaust with steam or water injection, a costly process because of the large amounts of highly treated water required. NO_x concentrations range from 35 to 70 ppm for gas-fired units, and 50 to 150 ppm for oil-fired units when steam injection is used. SCR is being applied to remove an additional 60-85% of the NO_x.

In a combined-cycle plant, exhaust gas is cooled by a superheater and pre-evaporator to between 540F and 750F before it is subjected to SCR. Further heat recovery is accomplished after the SCR reactor.

SCR requires a large amount of catalyst in this application because the gas volume is relatively large. Also the catalyst must be resistant to thermal shock because the gas temperature may change rapidly when operating load changes. On the other hand, a highly reactive, efficient catalyst can be used.

Tokyo Electric Power Corp operates a 1000-MW combined-cycle plant at the Futtsu station equipped with SCR. Another 1000-MW plant is under construction. Each plant consists of seven steam- and gas-turbine combinations, rated at 143 MW. NO_x emissions are reduced to 40-50 ppm with steam injection on a 15%-O_2 basis. SCR reduces them even further to 4-5 ppm using an ammonia-to-NO_x ratio of 1.0. The unreacted ammonia concentration is maintained below the outlet NO_x concentration.

In the clean-gas situation, it is expected that the catalyst can maintain 90% NO_x-removal efficiency even after 35,000 hr of operation.

Table 3: SCR-unit performance data for major utility coal-fired plants

Plant site	Unit No.	Boiler capacity, MW	Catalyst type	Space velocity, 1/hr	Inlet NO_x content, ppm	Outlet NO_x content, ppm	NO_x-removal efficiency, %	Unreacted ammonia, ppm	Inlet SO_2 content, ppm	Flyash content, lb/ft³	Comments
Takehara	1	½ x 250	Plate	2300	350	67	81	4	—	—	—
	2	½ x 250	Tube	2000	350	67	81	4	—	—	—
	3	700	Plate	2370	250	48	81	2	700	—[1]	—
Shimonoseki	1	175	H[2]	3000	420	180	57	1	1000	0.001	One of four catalyst layers replaced in 1984. One layer added in 1985
Shin-Ube	3	156	H[2]	4000	400	140	65	2	—	—	—
	—	125	H[2]	—	—	—	—	—	600	0.0009	One layer added in 1985
Mizushima	2	156	Plate	2200	350	120	66	2	—	—	—
Saijo	1	156	H[2]	4000	380	130	65	1	—	—	—
	2	250	H[2]	—	330	90	70	1	—	—	—

[1]Negligible—Unit has hot-side ESP [2]Honeycomb

NO_x concentration is higher, allowing a less reactive catalyst to be used, one with lower oxidation potential.

Two types of SCR systems for coal-fired boilers are the low-dust and high-dust systems. In the former, the flue gas is treated in a hot-side ESP upstream of the SCR. In the latter, the flue gas is treated by SCR ahead of a cold-side ESP. Both have advantages and disadvantages Because the optimum temperature for the SCR reactions is 570-750F, SCR is usually applied upstream of the FGD system at the economizer outlet.

In the low-dust system, catalyst erosion rarely occurs. However, the dust exiting the hot-side ESP is fine, relatively alkaline ash which tends to adhere to the catalyst surface. Ammonium bisulfate tends to deposit in the air heater.

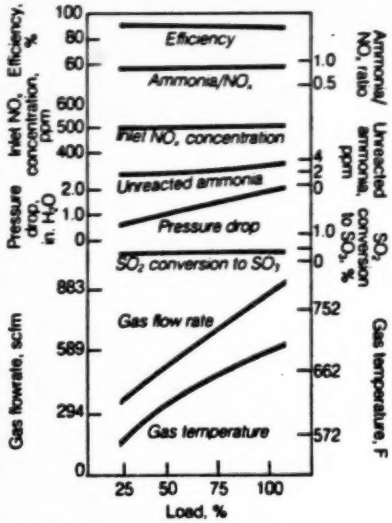
In the high-dust system, most of the bisulfate deposits on the ash, reducing the burden on the air heater. But if the flue gas is rich in unreacted ammonia, the flyash captured by the cold-side ESP may present disposal and utilization concerns. Note that this problem is not likely to occur if the ammonia concentration is kept below 5 ppm. Erosion of the catalyst can be prevented by selecting a hard catalyst and a dummy spacer and ensuring moderate gas velocity through the catalyst structure.

Although both high-dust and low-dust systems are feasible if the concentration of untreated ammonia is kept below 3 ppm, the low-dust system is far more popular.

Performance of a utility-boiler SCR unit during load changes is especially important (Fig 3). When the load is reduced, the flue-gas temperature drops but the NO_x-removal efficiency usually increases slightly. This happens because the gas volume decreases resulting in a longer residence time in the catalyst structure. By contrast, the SO_2-oxidation ratio will usually decrease at the lower temperature.

Also when temperature is reduced ammonium bisulfate may deposit on the catalyst. This usually occurs when temperature is in the 570-625F range, or about 480F with an air heater. Bisulfate formation on the catalyst at the higher temperature may be caused by the presence of a high concentration of SO_3 converted from SO_2, and high ammonia concentration in the reactor. Therefore, SCR plants for coal-fired boilers usually have an economizer-bypass line to maintain the gas temperature above 580F in the SCR reactor at reduced load. When the bypass is used, less heat is recovered in the economizer, but this is made up by recovering it in the downstream air heater instead.

SCR economic studies show that the major component of operating costs is catalyst life, usually based on a one-year replacement cycle for coal-fired-boiler applications. Recent data from Japanese powerplants suggest this may be much too conservative, that catalyst life is on the order of four to five years.



3. Turndown has a slight effect on the performance of the SCR unit

At the 700-MW Takehara unit, for example, ammonia slip has increased only slightly after three years. Ammonia slip, or the amount of injected ammonia that passes through unreacted, is an indicator of when catalyst should be replaced or reinforced.

In general, the performance of catalyst degrades because of: 1) sintering, or a reduction of pore volume; 2) blinding of pores by solid deposits; 3) poisoning by alkali compounds, especially potassium; 4) poisoning by SO_3; and 5) erosion by flyash. In order to maintain NO_x-removal efficiency, the NH_3 injection rate is increased over time, which increases the likelihood of a rise in ammonia slip as well. This, in turn, causes more ammonium bisulfate to deposit in the air heater. Eventually, the catalyst has to be replaced or a new layer added.

SCR/FGD in tandem

All Japanese coal-fired boilers with SCR units have gypsum-producing FGD plants downstream. The SCR unit can have some impact on the FGD unit.

One problem involves the ammonia contained in the wastewater from the FGD plant. FGD wastewater contains nitrogen compounds derived from the NO_x in the flue gas. With SCR, the ammonia content will increase but the amount of other nitrogen-bearing compounds decreases because the NO_x has been removed. Thus, the net amount of nitrogen-bearing compounds may not increase appreciably, but the ammonia content may have to be treated through biological or other means.

Of more importance to the FGD system is plugging of the regenerative reheater. The oxidation of SO_2 to SO_3 by the SCR catalyst tends to increase deposit formation. The reheater can usually be operated continuously for a year.

Edited by Jason Makansi